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Our Reference: UMJ-102-B (UM1544p1) PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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Applicant:	Curtis L. Munson & Ralph T. Yang
Serial Number:	09/632,891
Filing Date:	August 7, 2000
Examiner/Art Group Unit:	Tam M. Nguyen/1764
Title:	SULFUR RESISTANT ADSORBENTS

CERTIFICATION OF MAILING AND TRANSMITTAL LETTER

Sir:

Transmitted with this document is a revised Appeal Brief in triplicate in the above-identified application.

X No additional fee is required.

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I hereby certify that this correspondence is being deposited with the United States Postal Service as First Class Mail in an envelope addressed to: Assistant Commissioner of Patents, Washington, D.C. 20231, on **November 25, 2002.**

Julia Church Dierker
Attorney for Applicant(s)
Registration No. 33368
(248) 649-9900

DIERKER & GLASSMEYER, P.C.
3331 West Big Beaver Rd., Suite 109
Troy, Michigan 48084-2813

JCD/jc

3/3

Our Reference: UMJ-102-B (UM-1544p1) PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

Appellants:	Curtis L. Munson & Ralph T. Yang
Serial Number:	09/632,891
Filing Date:	August 7, 2000
Examiner/Art Group Unit:	Tam M. Nguyen/1764
Title:	SULFUR RESISTANT ADSORBENTS

APPEAL BRIEF

Assistant Commissioner of Patents
Washington, D.C. 20231

Sir:

Please enter the following revised Appeal Brief in the appeal filed July 8, 2002, and in response to the Notification of Non-Compliance with 37 C.F.R. 1.192(c) mailed October 23, 2002.

REAL PARTY IN INTEREST

The Assignees, the Regents of the University of Michigan and Chevron U.S.A., are the real parties in interest.

RELATED APPEALS AND INTERFERENCES

None.

STATUS OF CLAIMS

Claims 1-25 and 27-65 are pending and are attached as the Appendix. Claims 1-14, 27-30, 32, 33 and 65 have been rejected. Claims 15-25 and 31 have been allowed. **Claims 34-64 have no specific grounds of rejection set forth in the Final Office action, and it is thus assumed that they are allowed.**

STATUS OF AMENDMENTS

An Amendment under Rule 37 C.F.R. § 1.116 was filed on Septebmer 9, 2002 and was not entered by the Examiner.

INTRODUCTION

Appellants believe that an important argument has not been adequately and fully considered by the Examiner during prosecution of the above-identified application. In his "Response to Arguments," the Examiner asserts that Appellants have not shown why it is not obvious to use a feed which contains a small amount (the Examiner's example of "small" is 0.01 ppm or 1 ppb) of hydrogen sulfide (his point being that then one would not need Appellants' hydrogen sulfide resistant adsorbents). On the contrary, Appellants certainly have shown why this is not obvious--in the specification itself and in two responses, Appellants have specifically shown that it is highly cost prohibitive and impractical to desulfurize gas streams to the level suggested by the Examiner. Appellants will show in detail hereinbelow that the Examiner's grounds for rejection is flawed.

Further, the Examiner stated that claims 15-25 and 31 are allowed. In the Examiner's statement of reasons for the indication of allowable subject matter, he stated that "essentially all" cationic sites of the ion-exchanged zeolite contain silver cation or copper cation "as called for in claim 15". However, Appellants again, as they did in two responses, call the Examiner's attention to claim 15 as originally filed (and as it remains), which recites that "**a majority**" of the sites have silver cation or copper cation present. The "essentially all" language to which the Examiner refers is recited in dependent claim 23. Appellants note the allowance of claims 15-25 and 31; however, they request that the Examiner amend his reasons for allowance.

SUMMARY OF THE INVENTION

The present invention relates generally to adsorbents for selectively separating olefins from paraffins, and to adsorbents for selectively separating dienes from a mixture, particularly one containing mono-olefins. More particularly, the

present invention relates to such adsorbents which are resistant to and/or tolerant of hydrogen sulfide poisoning. (Page 1, lines 11-17)

The presence of sulfur compounds is generally known to have deleterious effects in various petroleum refining processes. For example, in catalytic reforming, the catalyst used is often platinum supported on high-purity alumina. However, the platinum on the catalyst is seriously deactivated by sulfur compounds, and therefore the feedstock is desulfurized to less than 3 ppm by weight of sulfur before the reforming. See, for example, Tedder, J.M., A. Nechvatal and A.H. Jubb, Basic Organic Chemistry, Part 5: Industrial Products, Chapter 3.2, "Preparation of Primary Petrochemicals: Reforming," pp. 53-60 (1975).

U.S. Patent No. 6,063,723 issued to Miller also demonstrates that sulfur is known to have deleterious effects. The '723 patent discloses a sulfur tolerant zeolite catalyst for use in catalytic dehydrogenation of light paraffinic hydrocarbons. This catalyst is prepared such that it has a very specific alkali to aluminum ratio in the zeolite. Although this patent's disclosed sulfur tolerance is better than that of previously known catalysts, it is still only disclosed to be tolerant of sulfur in the range of up to 2 ppm sulfur, and more preferably up to 0.5 ppm sulfur.

Cracked gas streams include a blend of many components, including olefins, paraffins, H_2, H_2S, etc. The cracked gas streams are first subjected to caustic scrubbing in order to remove hydrogen sulfide. Then, it is necessary to separate olefins from paraffins in these streams in order to obtain a product rating. Conventional methods for such separation include multiple distillation runs, one of which removes, for example, H_2 and methane. Thereafter, the stream includes C_2's and higher, mixed with olefins and paraffins. This stream is then distilled to separate the olefins from the paraffins. However, this distillation is very energy intensive and difficult to accomplish because of relatively close volatilities when alkene\alkane (olefin\paraffin) separation is required. For example, ethane\ethylene separation is carried out at about -25°C and 320 psig (2.603 MPa) in a column containing over 100 trays, and propane\propylene separation is performed by an equally energy-intensive distillation at about 38°C and 260 psig (1.8 MPa). It is evident that high

capital costs and high operational costs are incurred in any cryogenic distillation approach.

Further, it is also very difficult to separate a diene from a mixture, particularly one containing mono-olefin. Purifying the mono-olefin, 1-butene, is particularly troublesome due to the closeness of its boiling point to that of 1,3-butadiene. In order to increase the purity of 1-butene, it is necessary to separate it from other hydrocarbons. Ordinarily, fractionation alone is cost prohibitive for completely separating 1,3-butadiene to achieve the desired purity of 1-butene in these mixtures. Presently, butadiene is separated from olefins and paraffins primarily by distillation with selective solvents and by absorption using solutions of absorbents. Extractive distillation is relatively energy-intensive, complex and not economical. There are several drawbacks, in that this puts water vapor and/or solvent vapor into the regenerated product. Further, metal salts often are instable in solution and will precipitate out of solution. Selective absorption with metal salt solution involves additional stages with recycling of streams between stages to remove water vapor and/or solvent vapor from the regenerated product. This method has the disadvantage of being energy-intensive and requiring handling and recirculating of solvent streams which themselves contain contaminants or are subject to degradation. Current processes for olefin\paraffin separation have not been sufficiently selective to economically achieve the desired result for purifying mixtures of unsaturated hydrocarbons.

Thus, it is an object of the present invention to provide adsorbents which would cost-effectively and successfully achieve separation of olefins from paraffins. It is a further object of the present invention to provide adsorbents which would cost-effectively and successfully achieve separation of dienes from mono-olefins. Still further, it is an object of the present invention to provide such adsorbents which are advantageously resistant to and/or tolerant of sulfur poisoning. (Page 3, line 3 - Page 5, line 12)

A method of separating gaseous alkene selected from the group consisting of ethylene, propylene and mixtures thereof, from a gaseous mixture including the alkene and hydrogen sulfide, comprises the steps of:

contacting the gaseous mixture with an adsorbent which preferentially adsorbs the alkene, at a selected temperature and pressure, thereby producing a non-adsorbed component and an alkene-rich adsorbed component; the adsorbent comprising a carrier having a surface area, the carrier having a monolayer of a silver compound dispersed on substantially the entire surface area, the silver compound releasably retaining the alkene; and the carrier comprising a plurality of pores having a pore size greater than the effective molecular diameter of the alkene; and

changing at least one of the pressure and temperature to thereby release the alkene-rich component from the adsorbent;

wherein the adsorbent substantially maintains its adsorbent capacity and preference for the alkene when the hydrogen sulfide is present in amounts greater than about 3 ppm. (Page 8, line 6; Page 8, line 17 - Page 9, line 24)

A method for separating a diene from a mixture including the diene and a sulfur compound, comprises the step of:

contacting the mixture with an adsorbent which preferentially adsorbs the diene, at a selected temperature and pressure, thereby producing a non-adsorbed component and a diene-rich adsorbed component, wherein the adsorbent comprises an ion-exchanged zeolite selected from the group consisting of zeolite X, zeolite Y, zeolite LSX, and mixtures thereof, the zeolite having exchangeable cationic sites, and a majority of the sites having silver cation or copper cation present, and wherein the preferential adsorption occurs by π-complexation, and further wherein the adsorbent substantially maintains its adsorbent capacity and preference for the diene when the sulfur compound is present in amounts greater than about 3 ppm. (Page 11, lines 1-17)

ISSUES ON APPEAL

1. Whether claims 1-25 and 27-65 are patentable over Ogawa U.S. Patent No. 6,042,797 in view of Yang, R.T. and E.S. Kikkinides, "New Sorbents for Olefin/Paraffin Separations by Adsorption via π-Complexation," AIChE Journal, Vol. 41, No. 3 (March 1995) (AIChE-1995), further in view of Ramachandran et al., U.S. Patent No. 5,744,687 ('687) under 35 U.S.C. § 103(a); or

alternately over Milton U.S. Patent No. 2,882,243, when none of the references teach or suggest a sorbent which is resistant to hydrogen sulfide?

2. Whether claim 10 is patentable over Ogawa U.S. Patent No. 6,042,797 in view of Yang, R.T. and E.S. Kikkinides, "New Sorbents for Olefin/Paraffin Separations by Adsorption via π-Complexation," AIChE Journal, Vol. 41, No. 3 (March 1995) (AIChE-1995) when Ogawa teaches away from π-Complexation and there is no suggestion or motivation to combine Ogawa with AIChE 1995?

3. Whether claims 2, 6, 7, 12 and 14 are patentable over Ogawa U.S. Patent No. 6,042,797 in view of Yang, R.T. and E.S. Kikkinides, "New Sorbents for Olefin/Paraffin Separations by Adsorption via π-Complexation," AIChE Journal, Vol. 41, No. 3 (March 1995) (AIChE-1995) when Ogawa uses a zeolite adsorbent, teaches away from π-Complexation, and there is no suggestion or motivation to combine Ogawa with AIChE 1995?

GROUPING OF CLAIMS

Claims 1-25 and 27-65 stand together. Claim 10 is separately patentable from any other claim. Claims 2, 6, 7, 12 and 14 are separately patentable from any other claim. Claims 29-32 and 64 are separately patentable from any other claim.

The specific reasons for the separate patentability of each group of claims is set forth in the argument section of this Appeal Brief.

ARGUMENT

The Examiner's Rejection

Claims 1-7, 10-14, 33 and 65 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Ogawa in view of Yang, R.T. and E.S. Kikkinides, "New Sorbents for Olefin/Paraffin Separations by Adsorption via π-Complexation," AIChE Journal, Vol. 41, No. 3 (March 1995) (AIChE-1995).

The Examiner states that Ogawa discloses a process for removing ethylene from a gas mixture containing ethylene and a sulfur compound (e.g., sulfur

oxide) by contacting the gas mixture with an adsorbent which comprises a silver compound (silver nitrate) and zeolite. The Examiner recognizes that Ogawa does not specifically disclose that the carrier comprises a monolayer of a silver compound on the adsorbent surface. However, he stated that it is optional that only silver compound is impregnated on the adsorbent. Therefore, the Examiner concluded that this limitation is embraced by the reference. The Examiner also recognized that the reference does not disclose that the retaining of the alkene is accomplished by formation of π-complexation bonds. However, the Examiner **assumes** that Ogawa at col. 2, line 59 through col. 8, line 7 shows that the bonds between the silver compound and alkene occur by π-complexation.

The Examiner notes that Ogawa does not disclose that silver compound is dispersed on the adsorbent, and does not disclose the silver compound is silver halide and the carrier is silica which has a surface area between 50 to 2,000 m^2/g.

The Examiner states that AIChE 1995 discloses a process for separating ethylene/propylene from a paraffinic feed. The Examiner states that the AIChE 1995 adsorbent comprises a **silica** support (although silica is **not** disclosed in the AIChE 1995 reference) and silver salt (e.g., $AgNO_3$, AgCl).

The Examiner concluded that it would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Ogawa process by using a feedstock which contains a "small amount (e.g. 0.01 ppm or 1 ppb) of hydrogen sulfide because it would be expected that the tiny amount of hydrogen sulfide present in the Ogawa feed would not affect the outcome of the Ogawa process." (Final Action at page 4, lines 5-7) Further, the Examiner stated that it would have been obvious to disperse the silver compound on the carrier as taught by AIChE because AIChE discloses that it is effective to remove alkene when using an adsorbent wherein a silver compound is dispersed on the adsorbent.

Claims 8, 9, 29 and 30 stand rejected under 35 U.S.C. 103(a) as being unpatentable over the references as applied to claims 1-7 and 10-14 above, and further in view of Ramachandran et al. (5,744,687). The Examiner states that Ogawa does not disclose the operating temperatures and pressures (Appellants take issue

with this statement). However, the Examiner stated that Ramachandran discloses an adsorbent comprising a support which is **ion exchanged** with silver and/or copper. The Examiner states that the desorption step is operated at a temperature from about 100°C to 350°C. The Examiner stated that it would have been obvious to the skilled artisan to have modified the Ogawa process by utilizing the adsorption and desorption operating conditions of Ramachandran because the Ramachandran conditions are effective to adsorb and desorb ethylene/propylene.

Claims 27, 28 and 32 stand rejected under 35 U.S.C. 103(b) as being unpatentable over Milton (2,882,243). The Examiner admitted that Milton does not disclose that sulfur compounds are contained in the feedstock. However, the Examiner stated that it would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Milton process by utilizing a feedstock containing a tiny amount of hydrogen sulfide (e.g., 0.01 ppm) because it would be expected that the tiny amount of hydrogen sulfide present in the feedstock of Milton would not affect the outcome of the process of Milton.

ISSUE 1. **Whether claims 1-25 and 27-65 are patentable over Ogawa U.S. Patent No. 6,042,797 in view of Yang, R.T. and E.S. Kikkinides, "New Sorbents for Olefin/Paraffin Separations by Adsorption via π-Complexation," AIChE Journal, Vol. 41, No. 3 (March 1995) (AIChE-1995), further in view of Ramachandran et al., U.S. Patent No. 5,744,687 ('687) under 35 U.S.C. § 103(a); or alternately over Milton U.S. Patent No. 2,882,243, when none of the references teach or suggest a sorbent which is resistant to hydrogen sulfide?**

Appellants answer: yes

Examiner answers: no

The Examiner asserts that "applicant has not shown why it is not obvious to use a feed which contains a small amount of hydrogen sulfide (as stated above, his examples of "small" are 0.01 ppm or 1 ppb). Moreover, the Examiner points out that the present specification at page 13, lines 13-22 states that a conventional cracked gas stream contains about 0.01 mole% hydrogen sulfide, and the Ogawa feed is a *conventional cracked gas*. Therefore, it would be expected that at least hydrogen sulfide is contained in the Ogawa feedstock." (emphasis added)

First, the Examiner did not quote the above cite from Appellants' specification in context. The full phrase reads that "a conventional cracked gas stream **before any desulfurizing distillation steps** contains hydrogen sulfide present in amounts of about 0.01 mole% [100 ppm]."

Second, Ogawa does **not** disclose that its feed is a "conventional cracked gas." In sharp contrast, Ogawa states throughout the specification that its gas is "**an exhaust gas discharged from an internal combustion engine**." (Ogawa at Col. 1, lines 8-9; Col. 3, lines 16-18; Col. 7, lines 1-5; etc.) As is known in the art and as stated in the Summary of the Invention above, conventional cracked gas streams are generally desulfurized to predetermined limits before further refining. Gasoline for internal combustion engines is an **end product** of petroleum refining; and exhaust gas contains byproducts from internal combustion. As is evident, Ogawa's **exhaust gas** is NOT a "conventional cracked gas stream," contrary to the assertion by the Examiner.

Appellants submit that the Examiner's assertion that the skilled artisan would just know to use a gas stream with a tiny amount of hydrogen sulfide begs the issue. Further, his assertion is also not practical. As stated in Applicants' specification at page 3, lines 5-10:

> [I]n catalytic reforming, the catalyst used is often platinum supported on high-purity alumina. However, the platinum on the catalyst is seriously deactivated by sulfur compounds, and therefore the feedstock is desulfurized to less than 3 ppm by weight of sulfur before the reforming.

In order to preserve the expensive platinum in a catalytic reforming process, the desulfurization is taken close to the limits--in other processes, one would generally not see feedstocks having as low as 3 ppm hydrogen sulfide. **As such, it would be extremely expensive and time consuming, if not impossible, to desulfurize feedstock to a level of "0.01 ppm" as suggested by the Examiner.**

Appellants' invention as defined in claims 1, 11, 15, 29, 30, 32, 34, 48 and 55 is predicated on the fact that <u>it is not necessary to desulfurize to extremely low levels</u>:

> [T]his novel adsorbent fortuitously and unexpectedly substantially maintains its adsorbent capacity and preference for the alkene in the presence of the sulfur compound. This is highly desirable, as this sulfur tolerant and/or resistant adsorbent (as well as the other novel adsorbents described hereinbelow) **obviates step(s) conventionally necessary to desulfurize the cracked gas stream(s)**. (emphasis added) page 10, lines 28-36

The Examiner is further directed to the instant application at page 13, lines 13-22:

> The novel adsorbents were exposed to very severe amounts, for example the hydrogen sulfide was present in amounts up to about 66 mole%. **In sharp contrast, a conventional cracked gas stream before any desulfurizing distillation steps contains hydrogen sulfide present in amounts of about 0.01 mole% [100 ppm].** As such, the data presented hereinbelow indicate that **the novel adsorbents of the present invention would be quite robust, ie. very tolerant and/or resistant to H_2S under normal operating conditions**. (emphasis added)

As Appellants have shown above that Ogawa does not teach or suggest a gas containing hydrogen sulfide, it is requested that it be removed as a primary reference. Ogawa states at Col. 6, lines 29 that "[t]he adsorbent is effective also in a case where the gas contains not only ethylene but also . . . sulfur oxide." In Ogawa, the olefins are being removed from the atmosphere or an exhaust gas. Exhaust gas contains by-products of burning: sulfur oxides, carbon oxides, etc. In Appellants' invention as defined in claims 1, 11, 15, 29, 30, 32, 34, 48 and 55, it is recited that the adsorbent substantially maintains its adsorbent capacity and selectivity in the presence of hydrogen sulfide. This is remarkable in that hydrogen sulfide has been recognized as being capable of potentially deactivating entire adsorbents. **Thus, hydrogen sulfide has been recognized as reacting detrimentally; *whereas sulfur oxide is substantially an inert by-product.***

Neither Ogawa nor any of the other cited references disclose adsorbents which substantially maintain their adsorbent capacity and selectivity in the presence of hydrogen sulfide as recited in Appellants' claims 1, 11, 15, 29, 30, 32, 34, 48 and 55. As such, it is submitted that Appellants' invention as defined in these claims, as well as in all claims dependent therefrom, is not anticipated, taught or

rendered obvious by Ogawa or AIChE 1995, either alone or in combination, and patentably defines over the art of record.

Applicants submit that the recitation in claims 29-32 and 64 of hydrogen sulfide present in amounts up to about 66 mole% renders these claims and any dependent therefrom allowable. None of the cited references teach or suggest adsorbents which can function in an atmosphere having up to about 66 mole% hydrogen sulfide present (as stated above, in fact they do not even mention hydrogen sulfide). As such, it is submitted that claims 29-32 and 64 are separately patentable from any other claim.

It is respectfully submitted that the Examiner has used impermissible hindsight from Applicants' disclosure in order to **assume** that Ogawa teaches or even suggests a gas stream having hydrogen sulfide therein. It is further submitted that, in doing so, the Examiner has made errors in logic and consequently in the technical field at hand.

The Federal Circuit has spoken to the issue of impermissible hindsight on numerous occasions. In *In re David H. Fine*, 837 F.2d 1071, 5 U.S.P.Q. (BNA) 1596 (Fed. Cir. 1988), the court stated:

> To reach a proper conclusion under § 103, the decisionmaker must step backward in time and into the shoes worn by [a person having ordinary skill in the art] when the invention was unknown and just before it was made. In light of *all* the evidence, the decisionmaker must then determine whether . . . the claimed invention as a whole would have been obvious at *that* time to *that* person. 35 U.S.C. § 103. The answer to that question partakes more of the nature of law than of fact, for it is an ultimate conclusion based on a foundation formed of all the probative facts. (emphasis in original) Id. at 1073-74, quoting *Panduit Corp. v. Dennison Mfg. Co.*, 810 F.2d 1561, 1566 (Fed. Cir. 1987)

> Further, the *Fine* court stated at 1075:

> Because neither Warnick nor Eads, alone or in combination, suggests the claimed invention, the Board erred in affirming the Examiner's conclusion that it would have been obvious to substitute the Warnick nitric oxide detector for the Eads sulfur dioxide detector in the Eads system. *ACS Hosp. Sys.*, 732 F.2d at 1575-77, 221 USPQ at 931-33. **The Eads and Warnick references disclose, at most, that one skilled in the art might find it obvious to try the claimed invention. But whether a particular combination might be**

> **"obvious to try" is not a legitimate test of patentability**. [references omitted] (emphasis added)

The first quote above from the *Fine* court makes it clear that the Examiner must "step backward in time" to just before the present invention was made, and look at Ogawa and AIChE-1995 **without knowledge gleaned from Appellants' disclosure**. If the Examiner did this, in light of the detailed discussion above of what the skilled artisan would glean from the reference, he would come to the conclusion that neither reference speaks to the issue of hydrogen sulfide at all.

The Examiner's assertion seems to be that the skilled artisan would find it "obvious to try" to run a gas stream having hydrogen sulfide therein over the sorbents of the references. "**But whether a particular combination might be 'obvious to try' is not a legitimate test of patentability**." Id. (emphasis added)

Obviousness is tested by what the combined teaching of the references would have suggested to those of ordinary skill in the art. See *In re Keller*, 642 F.2d 413, 425, 208 U.S.P.Q. (BNA) 871, 881 (CCPA 1981). Approaches to obviousness determinations which focus merely on identifying and tabulating missing elements in hindsight retrospect imbue one of ordinary skill in the art with knowledge of the invention in suit, when no prior art reference or references of record convey or suggest that knowledge (ie. assuming that Ogawa's sorbent would adequately function in the presence of hydrogen sulfide), and fall victim to the insidious effect of the hindsight syndrome wherein that which only the inventor taught is used against its teacher. (emphasis added). *W.L. Gore & Assoc. v. Garlock, Inc.*, 721 F.2d 1540, 1553, 220 U.S.P.Q. (BNA) 312, 313 (Fed. Cir. 1983).

For all the above reasons, it is submitted that Appellants' invention as defined in claims 1-25 and 27-65 is not anticipated, taught or rendered obvious by either of Ogawa or AIChE-1995, either alone or in combination, and patentably defines over the art of record.

It is further submitted that the arguments for patentability of the claims hereinabove and hereinbelow are intended to be cumulative.

ISSUE 2. Whether claim 10 is patentable over Ogawa U.S. Patent No. 6,042,797 in view of Yang, R.T. and E.S. Kikkinides, "New Sorbents for Olefin/Paraffin Separations by Adsorption via π-Complexation," AIChE Journal, Vol. 41, No. 3 (March 1995) (AIChE-1995) when Ogawa teaches away from π-Complexation and there is no suggestion or motivation to combine Ogawa with AIChE 1995?

Appellants answer: yes

Examiner answers: no

Ogawa does not ever mention π-Complexation, and in fact teaches away from bonding by π-Complexation. Ogawa states that the sorbent has a "strong power for holding the adsorbed hydrocarbon," Col 3, line 45-46, and that it desorbs "starting at" 200C to 300C. As stated in the specification, π-Complex bonds are relatively weak. Further, Appellants recite that desorption takes place between about 70C and about 200C. π-Complexation occurs with d-block transition metals, however, Ogawa states that any transition metals may be used (Col. 6, lines 6 et seq) This points away from Ogawa even contemplating π-Complexation bonding.

As such, the Examiner has offered no motivation to combine Ogawa with AIChE 1995. The Federal Circuit spoke to this issue in the recent case of In re Lee, 277 F.3d 1338, 61 U.S.P.Q.2d (BNA) 1430 (Fed. Cir. 2002):

> With respect to Lee's application, neither the examiner nor the Board adequately supported the selection and combination of the Nortrup and Thunderchopper references to render obvious that which Lee described. The examiner's **conclusory statements** that "the demonstration mode is just a programmable feature which can be used in many different devices for providing automatic introduction by adding the proper programming software" and that "another motivation would be that the automatic demonstration mode is user friendly and it functions as a tutorial" do not adequately address the issue of motivation to combine. **This factual question [*1344] of motivation is material to patentability, and could not be resolved on subjective belief and unknown authority**. It is improper, in determining whether a person of ordinary skill would have been led to this combination of references, simply to "[use] that which the inventor taught against its teacher." W.L. Gore v. Garlock, Inc., 721 F.2d 1540, 1553, 220 U.S.P.Q. (BNA) 303, 312-13 (Fed. Cir. 1983). Thus the Board must not only assure that the requisite findings are made, based on evidence of record, but must also explain the reasoning by which the findings are deemed to support, the agency's conclusion. (emphasis added)

It is submitted that the Examiner has not provided any evidence re: motivation to support combination of the two references. For this further reason, it is submitted that claim 10 is not anticipated, taught or rendered obvious by any of the cited references, and patentably defines over the art of record.

ISSUE 3. Whether claims 2, 6, 7, 12 and 14 are patentable over Ogawa U.S. Patent No. 6,042,797 in view of Yang, R.T. and E.S. Kikkinides, "New Sorbents for Olefin/Paraffin Separations by Adsorption via π-Complexation," <u>AIChE Journal</u>, Vol. 41, No. 3 (March 1995) (AIChE-1995) when Ogawa uses a zeolite adsorbent, teaches away from π-Complexation, and there is no suggestion or motivation to combine Ogawa with AIChE 1995?

Appellants answer: yes

Examiner answers: no

Ogawa uses a zeolite sorbent, and only states that silica may be optionally included as a binder. As stated above, Ogawa also teaches away from π-Complexation. Appellants' invention as defined in claims 2, 6, 7, 12 and 14 recites a silica carrier. Neither Ogawa nor AIChE 1995 recite a silica carrier. As such, it is submitted that there is no motivation to combine Ogawa with AIChE 1995; and even if such combination were made, it would not render a sorbent having silica as a carrier. For these reasons, it is submitted that Appellants' invention as defined in claims 2, 6, 7, 12 and 14 are not anticipated, taught or rendered obvious by any of the cited references, and patentably defines over the art of record.

REQUEST FOR INDICATION OF ALLOWANCE OF CLAIMS 34-64

The Examiner has set forth in the Final Office action no specific grounds of rejection of Appellants' invention as defined in claims 34-64. Appellants thus assume that claims 34-64 are allowed, notice of which is respectfully requested.

<u>CONCLUSION</u>

For the reasons stated above, it is submitted that there is no teaching or suggestion in either of Ogawa or AIChE-1995, of an adsorbent able to substantially maintain its capacity and selectivity in the presence of hydrogen sulfide. It is further submitted that the Examiner has not made his prima facie case of

obviousness with regard to the claims reciting silica as the carrier or π-complexation for bonding.

Thus, it is respectfully submitted that Appellants' invention as set forth in claims 1-25 and 27-65 patentably defines over the cited references and is not anticipated, taught or rendered obvious thereby. As such, it is respectfully submitted that the Examiner's final rejection of claims 1-14, 27-30, 32, 33 and 65 is erroneously based, and its reversal is respectfully requested.

No oral hearing is requested.

This revised Appeal Brief is being filed in triplicate, including one original and two copies.

Respectfully submitted,

DIERKER & GLASSMEYER, P.C.

Julia Church Dierker
Attorney for Appellants
Registration No. 33368
(248) 649-9900

3331 West Big Beaver Rd.
Suite 109
Troy, Michigan 48084-2813

Dated: November 25, 2002

APPENDIX

CLAIMS AT ISSUE IN APPEAL OF S.N. 09/632,891

1. A method of separating gaseous alkene selected from the group consisting of ethylene, propylene and mixtures thereof, from a gaseous mixture including the alkene and hydrogen sulfide, the method comprising the steps of:

contacting the gaseous mixture with an adsorbent which preferentially adsorbs the alkene, at a selected temperature and pressure, thereby producing a non-adsorbed component and an alkene-rich adsorbed component; the adsorbent comprising a carrier having a surface area, the carrier having a monolayer of a silver compound dispersed on substantially the entire surface area, the silver compound releasably retaining the alkene; and the carrier comprising a plurality of pores having a pore size greater than the effective molecular diameter of the alkene; and

changing at least one of the pressure and temperature to thereby release the alkene-rich component from the adsorbent;

wherein the adsorbent substantially maintains its adsorbent capacity and preference for the alkene in the presence of the hydrogen sulfide.

2. The method as defined in claim 1 wherein the silver compound is silver nitrate ($AgNO_3$) and the carrier is silica (SiO_2).

3. The method as defined in claim 1 wherein the silver compound is a silver salt, and wherein the salt is selected from the group consisting of acetate, benzoate, bromate, chlorate, perchlorate, chlorite, citrate, fluoride, nitrate, nitrite, sulfate, and mixtures thereof.

4. The method as defined in claim 1 wherein the carrier has a BET surface area greater than about 50 square meters per gram and up to about 2,000 square meters per gram, and comprises a plurality of pores having a pore size greater than about 3 angstroms and up to about 10 microns.

5. The method as defined in claim 1 wherein the carrier is a high surface area support selected from the group consisting of refractory inorganic oxide, molecular sieve, activated carbon, and mixtures thereof.

6. The method as defined in claim 5 wherein the refractory inorganic oxide is selected from the group consisting of pillared clay, alumina and silica.

7. The method as defined in claim 1 wherein the silver compound is a silver (I) halide and the carrier is silica.

8. The method as defined in claim 1 wherein the selected pressure of preferential adsorption is a first pressure, and the pressure of release is a second pressure less than the first pressure, and wherein the first pressure is in a range of about 1 atmosphere to about 35 atmospheres, and further wherein the second pressure is in a range of about 0.01 atm to about 5 atm.

9. The method as defined in claim 1 wherein the selected temperature of preferential adsorption is a first temperature, and the temperature of release is a second temperature greater than the first temperature, and wherein the first temperature is in a range of about O°C to about 50°C, and further wherein the second temperature is in a range of about 70°C to about 200°C.

10. The method as defined in claim 1 wherein the retaining of the alkene is accomplished by formation of π-complexation bonds between the silver compound and the alkene.

11. A sulfur tolerant adsorbent for preferential adsorption of gaseous alkene from a gaseous mixture including the alkene and hydrogen sulfide, the adsorbent comprising:

a carrier; and

a silver compound supported on the carrier, wherein the silver compound is a silver salt, and wherein the salt is selected from the group consisting of acetate, benzoate, bromate, chlorate, perchlorate, chlorite, citrate, fluoride, nitrate, nitrite, and sulfate;

and wherein the adsorbent substantially maintains its adsorbent capacity and preference for the alkene in the presence of the hydrogen sulfide.

12. The adsorbent as defined in claim 11 wherein the adsorbent comprises finely divided particles of silica (SiO_2) with silver nitrate ($AgNO_3$) dispersed on and supported on the particles.

13. The adsorbent as defined in claim 11 wherein the carrier is selected from the group consisting of refractory inorganic oxide, molecular sieve, and activated carbon in particle form.

14. The adsorbent as defined in claim 13 wherein the refractory oxide is selected from the group consisting of pillared clay, alumina and silica.

15. A method for separating a diene from a mixture including the diene and a sulfur compound, the method comprising the step of:

contacting the mixture with an adsorbent which preferentially adsorbs the diene, at a selected temperature and pressure, thereby producing a non-adsorbed component and a diene-rich adsorbed component, wherein the adsorbent comprises an ion-exchanged zeolite selected from the group consisting of zeolite X, zeolite Y, zeolite LSX, and mixtures thereof, the zeolite having exchangeable cationic sites, and a majority of the sites having silver cation or copper cation present, and wherein the preferential adsorption occurs by π-complexation, and further wherein the adsorbent substantially maintains its adsorbent capacity and preference for the diene in the presence of the sulfur compound.

16. The method as defined in claim 15 wherein the diene is selected from the group consisting of butadiene, hexadiene, octadiene and mixtures thereof, and wherein the method further comprises the step of changing at least one of the pressure and temperature to thereby release the diene-rich component from the adsorbent.

17. The method as defined in claim 15 wherein the diene is 1,3-butadiene, and wherein the mixture includes 1,3-butadiene and at least one other C_4 unsaturated compound.

18. The method as defined in claim 15 wherein the majority of the cationic sites of the ion-exchanged zeolite contain the silver cation.

19. The method as defined in claim 15 wherein the majority of the cationic sites of the ion-exchanged zeolite contain the copper cation.

20. The method as defined in claim 15 wherein the mixture comprises at least one mono-olefin having as many carbon atoms as the diene, wherein the diene is selected from the group consisting of butadiene, hexadiene, octadiene, and mixtures thereof; and wherein the mono-olefin is selected from the group consisting of butene, hexene, octene, and mixtures thereof.

21. The method as defined in claim 20 wherein the mono-olefin is butene and the diene is butadiene.

22. The method as defined in claim 20 wherein the mixture comprises the mono-olefin in a gaseous state and saturated with the diene.

23. The method as defined in claim 15 wherein essentially all cationic sites of the ion-exchanged zeolite contain the silver cation.

24. The method as defined in claim 16 wherein the selected pressure of preferential adsorption is a first pressure, and the pressure of release is a second pressure less than the first pressure, wherein the first pressure is in a range of about 1 atmosphere to about 35 atmospheres, and wherein the second pressure is in a range of about 0.01 atmosphere to about 5 atmospheres.

25. The method as defined in claim 16 wherein the selected temperature of preferential adsorption is a first temperature, and the temperature of release is a second temperature greater than the first temperature, wherein the first temperature is in a range of about O°C to about 150°C, and wherein the second temperature is in a range of about 70°C to about 250°C.

27. The method as defined in claim 32 wherein the diene is selected from the group consisting of butadiene, hexadiene, octadiene, and mixtures thereof.

28. The method as defined in claim 27 wherein a selected pressure of preferential adsorption is a first pressure, and a pressure of release is a second pressure less than the first pressure, wherein the first pressure is in a range of about 1 atmosphere to about 35 atmospheres, and wherein the second pressure is in a range of about 0.01 atmosphere to about 5 atmospheres.

29. A method of separating gaseous alkene selected from the group consisting of ethylene, propylene and mixtures thereof, from a gaseous mixture including the alkene and a sulfur compound, the method comprising the steps of:

contacting the gaseous mixture with an adsorbent which preferentially adsorbs the alkene, at a selected temperature and pressure, thereby producing a non-adsorbed component and an alkene-rich adsorbed component; the adsorbent comprising a carrier having a surface area, the carrier having a monolayer of a silver compound dispersed on substantially the entire surface area, the silver compound releasably retaining the alkene; and the carrier comprising a plurality of pores having a pore size greater than the effective molecular diameter of the alkene; and

changing at least one of the pressure and temperature to thereby release the alkene-rich component from the adsorbent;

wherein the adsorbent substantially maintains its adsorbent capacity and preference for the alkene in the presence of the sulfur compound, wherein the sulfur compound is hydrogen sulfide, and wherein the hydrogen sulfide is present in amounts up to about 66 mole%.

30. A sulfur tolerant adsorbent for preferential adsorption of gaseous alkene from a gaseous mixture including the alkene and a sulfur compound, the adsorbent comprising:

a carrier; and

a silver compound supported on the carrier, wherein the silver compound is a silver salt, and wherein the salt is selected from the group consisting

of acetate, benzoate, bromate, chlorate, perchlorate, chlorite, citrate, fluoride, nitrate, nitrite, and sulfate;

wherein the carrier has a BET surface area greater than about 50 square meters per gram and up to about 2,000 square meters per gram and comprising a plurality of pores having a pore size greater than about 3 angstroms and up to about 10 microns;

and wherein the adsorbent substantially maintains its adsorbent capacity and preference for the alkene in the presence of the sulfur compound, wherein the sulfur compound is hydrogen sulfide, and wherein the hydrogen sulfide is present in amounts up to about 66 mole%.

31. The method as defined in claim 15 wherein the sulfur compound is hydrogen sulfide, and wherein the hydrogen sulfide is present in amounts up to about 66 mole%.

32. A method for separating a diene from a mixture including the diene and a sulfur compound, the method comprising the steps of:

contacting the mixture with an adsorbent which preferentially adsorbs the diene at a first temperature, thereby producing a non-adsorbed component and a diene-rich adsorbed component, wherein the adsorbent comprises zeolite A having exchangeable cationic sites, a plurality of the zeolite A sites having an alkali metal cation or an alkaline earth metal cation present; and

releasing the diene-rich adsorbed component from the adsorbent by elevating the temperature to a second temperature which ranges between about 70°C and about 120°C;

wherein the adsorbent substantially maintains its adsorbent capacity and preference for the diene in the presence of the sulfur compound, wherein the sulfur compound is hydrogen sulfide, and wherein the hydrogen sulfide is present in amounts up to about 66 mole%.

33. The sulfur tolerant adsorbent as defined in claim 11 wherein the carrier has a BET surface area greater than about 50 square meters per gram and up to

about 2,000 square meters per gram and comprising a plurality of pores having a pore size greater than about 3 angstroms and up to about 10 microns.

34. A method for separating a diene from a mixture including the diene and hydrogen sulfide, the method comprising the step of:

contacting the mixture with an adsorbent which preferentially adsorbs the diene, at a selected temperature and pressure, thereby producing a non-adsorbed component and a diene-rich adsorbed component, wherein the adsorbent comprises an ion-exchanged zeolite selected from the group consisting of zeolite X, zeolite Y, zeolite LSX, and mixtures thereof, the zeolite having exchangeable cationic sites, and at least some of the sites having silver cation or copper cation present, and wherein the preferential adsorption occurs by π-complexation, and further wherein the adsorbent substantially maintains its adsorbent capacity and preference for the diene in the presence of the hydrogen sulfide.

35. The method as defined in claim 34 wherein the diene is selected from the group consisting of butadiene, hexadiene, octadiene and mixtures thereof, and wherein the method further comprises the step of changing at least one of the pressure and temperature to thereby release the diene-rich component from the adsorbent.

36. The method as defined in claim 34 wherein the diene is 1,3-butadiene, and wherein the mixture includes 1,3-butadiene and at least one other C_4 unsaturated compound.

37. The method as defined in claim 34 wherein a majority of the cationic sites of the ion-exchanged zeolite contain the silver cation.

38. The method as defined in claim 34 wherein a majority of the cationic sites of the ion-exchanged zeolite contain the copper cation.

39. The method as defined in claim 34 wherein the mixture comprises at least one mono-olefin having as many carbon atoms as the diene, wherein the

diene is selected from the group consisting of butadiene, hexadiene, octadiene, and mixtures thereof; and wherein the mono-olefin is selected from the group consisting of butene, hexene, octene, and mixtures thereof.

40. The method as defined in claim 39 wherein the mono-olefin is butene and the diene is butadiene.

41. The method as defined in claim 39 wherein the mixture comprises the mono-olefin in a gaseous state and saturated with the diene.

42. The method as defined in claim 34 wherein essentially all cationic sites of the ion-exchanged zeolite contain the silver cation.

43. The method as defined in claim 35 wherein the selected pressure of preferential adsorption is a first pressure, and the pressure of release is a second pressure less than the first pressure, wherein the first pressure is in a range of about 1 atmosphere to about 35 atmospheres, and wherein the second pressure is in a range of about 0.01 atmosphere to about 5 atmospheres.

44. The method as defined in claim 35 wherein the selected temperature of preferential adsorption is a first temperature, and the temperature of release is a second temperature greater than the first temperature, wherein the first temperature is in a range of about O°C to about 150°C, and wherein the second temperature is in a range of about 70°C to about 250°C.

45. The method as defined in claim 34 wherein the at least some of the sites have silver cation present.

46. The method as defined in claim 34 wherein the at least some of the sites have copper cation present.

47. The method as defined in claim 34 wherein essentially all cationic sites of the ion-exchanged zeolite contain the copper cation.

48. A method for separating a diene from a mixture including the diene and hydrogen sulfide, wherein the diene is selected from the group consisting of butadiene, hexadiene, octadiene and mixtures thereof, wherein the mixture comprises at least one mono-olefin having as many carbon atoms as the diene, and wherein the mono-olefin is selected from the group consisting of butene, hexene, octene, and mixtures thereof, the method comprising the steps of:

contacting the mixture with an adsorbent which preferentially adsorbs the diene, at a selected temperature and pressure, thereby producing a non-adsorbed component and a diene-rich adsorbed component, wherein the adsorbent comprises an ion-exchanged zeolite selected from the group consisting of zeolite X, zeolite Y, zeolite LSX, and mixtures thereof, the zeolite having exchangeable cationic sites, and at least some of the sites having silver cation or copper cation present, and wherein the preferential adsorption occurs by π-complexation, and further wherein the adsorbent substantially maintains its adsorbent capacity and preference for the diene in the presence of the hydrogen sulfide;

changing at least one of the pressure and temperature to thereby release the diene-rich component from the adsorbent, wherein the selected pressure of preferential adsorption is a first pressure, and the pressure of release is a second pressure less than the first pressure, wherein the first pressure is in a range of about 1 atmosphere to about 35 atmospheres, and wherein the second pressure is in a range of about 0.01 atmosphere to about 5 atmospheres;

and wherein the selected temperature of preferential adsorption is a first temperature, and the temperature of release is a second temperature greater than the first temperature, wherein the first temperature is in a range of about O°C to about 150°C, and wherein the second temperature is in a range of about 70°C to about 250°C.

49. The method as defined in claim 48 wherein the mono-olefin is butene and the diene is butadiene.

50. The method as defined in claim 48 wherein the mixture comprises the mono-olefin in a gaseous state and saturated with the diene.

51. The method as defined in claim 48 wherein a majority of the sites have silver cation present.

52. The method as defined in claim 48 wherein a majority of the sites have copper cation present.

53. The method as defined in claim 48 wherein essentially all cationic sites of the ion-exchanged zeolite contain the copper cation.

54. The method as defined in claim 48 wherein essentially all cationic sites of the ion-exchanged zeolite contain the silver cation.

55. A sulfur tolerant adsorbent for preferential adsorption of gaseous diene from a gaseous mixture including the diene and hydrogen sulfide, the adsorbent comprising:

an ion-exchanged zeolite selected from the group consisting of zeolite X, zeolite Y, zeolite LSX, and mixtures thereof, the zeolite having exchangeable cationic sites, and at least some of the sites having silver cation or copper cation present, wherein the preferential adsorption occurs by π-complexation; and

wherein the adsorbent substantially maintains its adsorbent capacity and preference for the diene in the presence of the hydrogen sulfide.

56. The sulfur tolerant adsorbent as defined in claim 55 wherein the diene is selected from the group consisting of butadiene, hexadiene, octadiene and mixtures thereof.

57. The sulfur tolerant adsorbent as defined in claim 55 wherein the diene is 1,3-butadiene, and wherein the mixture includes 1,3-butadiene and at least one other C_4 unsaturated compound.

58. The sulfur tolerant adsorbent as defined in claim 55 wherein a majority of the cationic sites of the ion-exchanged zeolite contain the silver cation.

59. The sulfur tolerant adsorbent as defined in claim 55 wherein a majority of the cationic sites of the ion-exchanged zeolite contain the copper cation.

60. The sulfur tolerant adsorbent as defined in claim 55 wherein essentially all cationic sites of the ion-exchanged zeolite contain the silver cation.

61. The method as defined in claim 55 wherein essentially all cationic sites of the ion-exchanged zeolite contain the copper cation.

62. The sulfur tolerant adsorbent as defined in claim 55 wherein the mixture comprises at least one mono-olefin having as many carbon atoms as the diene, wherein the diene is selected from the group consisting of butadiene, hexadiene, octadiene, and mixtures thereof; and wherein the mono-olefin is selected from the group consisting of butene, hexene, octene, and mixtures thereof.

63. The sulfur tolerant adsorbent as defined in claim 62 wherein the mono-olefin is butene and the diene is butadiene.

64. The method as defined in claim 55 wherein the hydrogen sulfide is present in amounts up to about 66 mole%.

65. The method as defined in claim 1 wherein the gaseous mixture is contained in a conventional cracked gas stream before any desulfurizing distillation steps.